Proposal for *in situ* Enhancement of Electron Spin Polarization in Semiconductors.

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An extension of the original Overhauser effect to a more general nonequilibrium state was proposed by G. Feher, and demonstrated by Clark and Feher some forty years ago. It is suggested here that it might be possible to produce excess electron spin polarization by allowing the role of the nuclei to be played by other magnetic entities, such as paramagnetic impurities or adjacent magnetically ordered structures.

I. Background: Plans to utilize the spin degree of freedom of electrons (rather than only their charge) in the construction of semiconductor devices depend on the creation of a degree of spin polarization well in excess of the very small net alignment available in ordinary magnetic fields, especially at room temperature. An appealing way to achieve large polarization is to inject electrons from the majority Fermi sea in a ferromagnetic metal into the semiconductor. Giant magnetoresistance heterostructures, involving injection of the polarized electrons into, and transmission through, a nonmagnetic metal, raise hopes that such injection, and subsequent transport, will be possible in semiconductors also. So far, only a few successes have been claimed. These either involve cryogenic temperatures, or else use semiconducting material not favored in applications[1]. Also, in a certain semiconductor, anomalously high $g-$ values due to band structure effects have been reported[2],obviating the need for polarized injection, but, again, very low temperatures are needed. Some success has been reported involving optical methods, and probably reflection (rather than injection) from, a ferromagnet[3] .

Here, a method is proposed that avoids the need for injection or reflection altogether, and should function at room temperature. It is based on a kind of inversion of a generalized Overhauser effect proposed in 1959 by G. Feher[4], and subsequently realized experimentally by W. G. Clark and G. Feher[5]. The original Overhauser effect described greatly enhanced nuclear spin polarization resulting from strong excitation of the paramagnetic resonance of electrons in hyperfine interaction with the nuclei. The major insight achieved by Feher[4] was that the crucial feature of the Overhauser

effect was not the microwave excitation of the electrons, but simply their non-equilibrium distribution , no matter how produced. In the Clark-Feher experiment, the electrons are thrown out of equilibrium by an electric field applied to the semiconductor, (indium antimonide), heating the electrons to a temperature T_H, say. The hyperfine coupling of such a 'hot' electrons to the In^{115} nuclei causes a simultaneous spin flip of the electronic and nuclear spins. Single spin flips of the electron by spin-orbit coupling to the lattice also occur, but may be ignored initially. Single spin flips of the nuclei lead to very long spin lifetimes and may be ignored altogether. Let A_+, A_- denote the concentrations of upspin and downspin hot electrons, and B_+, B_- the number of upspin and downspin nuclei respectively. Then (since single spin flip processes are ignored for now), the master equation for A_+ reads, in the steady state

$$W_{-+\Rightarrow+-}A_-B_+ - W_{+-\Rightarrow-+}A_+B_- = 0 \tag{2}$$

where $W_{-+\Rightarrow+-}$ is the rate of simultaneous spin reversal of electron and nucleus, the former from down to up, the latter from up to down, and similarly for $W_{+-\Rightarrow-+}$. Since this mutual spin flip process, which does *not* conserve energy, is powered by the hot electrons, the ratio of the two W 's has a value appropriate to detailed balance at temperature T_H :

$$\frac{W_{-+\Rightarrow+-}}{W_{+-\Rightarrow-+}} = \exp\frac{2(\mu_A - \mu_B)H}{k_bT_H} \tag{3}$$

where μ_A, μ_B are the magnetic moments of the electrons and nuclei, and H the applied magnetic field. In this open, non-equilibrium system, the occupation numbers $A_\pm, B_\pm$ will be given by Boltzmann factors $e^{\pm\mu_AH/k_bT_A}$, $e^{\pm\mu_BH/k_bT_B}$ with their own temperatures T_A and T_B.Thus from equations (1) and (2),

$$\begin{aligned}\frac{A_+}{A_-}\frac{B_-}{B_+} &= e^{2\mu_AH/k_bT_A}e^{-2\mu_BH/k_bT_B} \\ &= e^{\frac{2(\mu_A-\mu_B)H}{k_bT_H}}\end{aligned} \tag{4}$$

whence

$$\frac{\mu_B}{T_B} = \frac{\mu_A}{T_A} - \frac{\mu_A - \mu_B}{T_H} \tag{5}$$

This shows that for thermal energies of the hot electron far in excess of the magnetic energies, $\frac{\mu_{A,B}H}{k_bT_{A,B}}$, the quantity determining the extent of nuclear polarization approaches $\frac{\mu_AH}{k_bT_A}$, which determines the much bigger electronic

polarization. In other words, $\frac{B_+}{B_-} \to \frac{A_+}{A_-}$ as $T_H \to \infty$. (Usually, this result is written as a greatly reduced effective nuclear temperature $T_B = \left(\frac{\mu_B}{\mu_A}\right) T_A$). Note that this assumes that the electron variables are 'robust', with T_A rigidly fixed. Equation (4) could equally well slave T_A to a rigidly fixed nuclear temperature. This difficulty is resolved by taking single flip processes into account (see next section)

II. This Proposal: The essence of this proposal is to let the role of the nuclei be assumed by magnetic entities (for example paramagnetic impurities with effective magnetic moments μ_B much larger than the electronic magnetic moment μ_A . Then $\mu_A \to \mu_B \left(\frac{T_A}{T_B}\right)$ as $T_H \to \infty$. In as much as in such a system $\frac{T_A}{T_B}$ might be of order one, the electrons will have acquired the magnetic moment of the impurity.

When single flip processes of the electrons and of the 'impurities' are not neglected, it is found that this result retains its validity if a certain inequality is satisfied. Note that the total concentrations of the A and B species scale out of equation (1) which is homogeneous of degree 2. Inclusion of single flip processes spoils the homogeneity and results in a concentration dependence. Writing $A_+ = A\cos^2\theta_A,\ A_- = A\sin^2\theta_A; B_+ = B\cos^2\theta_B,\ B_- = B\sin^2\theta_B$, with total concentrations A, B,the steady state master equations for A_+, B_+ now read

$$\begin{aligned} U + Aw^A_{+\to-}\cos^2\theta_A - Aw^A_{-\to+}\sin^2\theta_A &= 0 \\ -U + Bw^B_{+\to-}\cos^2\theta_B - Bw^B_{-\to+}\sin^2\theta_B &= 0 \end{aligned} \tag{6}$$

where

$$U = AB\left(W_{+-\Rightarrow-+}\cos^2\theta_A\sin^2\theta_B - W_{-+\Rightarrow+-}\sin^2\theta_A\cos^2\theta_B\right) \tag{7}$$

Here, $w^A_{+\to-} = w^A\exp(-\mu_A H/k_b T_A)$, $w^A_{-\to+} = w^A\exp(\mu_A H/k_b T_B)$ are the single flip rates for the electrons, and similarly for the 'impurities'. These flips are powered by lattice vibrations via spin-orbit coupling, so that both $T_A \& T_B$ are presumably of order of the lattice temperature. Although equations (5) and (6) can be reduced to a single quadratic, (for $\cos 2\theta_A$, for example), the coefficients are very involved. However, there appears to be one particularly simple solution for which $\theta_A = \theta_B = \theta$. If $U \neq 0$, this solution, according to equations (5), must satisfy

$$Aw^A_{+\to-}\cos^2\theta - Aw^A_{-\to+}\sin^2\theta = -(Bw^B_{+\to-}\cos^2\theta - Bw^B_{-\to+}\sin^2\theta) \tag{8}$$

or

$$\cot^2\theta = \frac{Aw^A_{-\to+} + Bw^B_{-\to+}}{Aw^A_{+\to-} + Bw^B_{+\to-}} \quad (9)$$

$$= \frac{1 + \frac{Bw^B}{Aw^A} e^{(\mu_B/T_B\ -\mu_A/T_A)H/k_b}}{1 + \frac{Bw^B}{Aw^A} e^{-(\mu_B/T_B\ -\mu_A/T_A)H/k_b}} \quad (10)$$

This is consistent with the earlier , concentration independent result $\cot^2\theta = e^{2\mu_B/k_bT_B}$ for very large T_H, provided $\frac{Bw^B}{Aw^A}e^{-(\mu_B/T_B\ -\mu_A/T_A)H/k_b}$ is much greater than 1, and $\mu_A/T_A << \mu_B/T_B$. (Note that, if T_H were allowed to go to infinity,i.e. $U \to 0$, at the beginning of the calculation, this solution would fail.). An improved solution may be obtained by writing $\theta_A = \theta + \delta_A$, $\theta_B = \theta + \delta_B$ in equation (5), in the definition (6) for U, and expanding to first order in the δ 's, resulting in two first order linear simultaneous equations for δ_A and δ_B.

III. Possible Implementation. In the above, the 'impurity' was characterized as a simple magnetic moment μ_B and its Zeemann energy in a magnetic field. To significantly enhance the electron spin polarization, the implanted impurity must have a large spin and/or an anomalously large $g-$ factor. In a magnetic field of 1 T, the electron polarization $(A_+/A_-) - 1$ in the absence of the impurity would only be 0.6% at room temperature. If the implant has a spin of 2, this figure would be increased to about 2.4%. (Although the analysis in section II was phrased in terms of an impurity with spin 1/2, the results are easily shown to hold for larger spins also, as long as the levels are equispaced). A further increase could come if spin-orbit coupling to the crystal lattice results in a large axial anisotropy energy for that implanted ion. This might conceivably amount to an additional effective field of one Tessler, giving 4.8% excess polarization. However, much better results can be obtained if the 'impurity' is replaced by be any magnetic structure with a lowest magnetic excitation energy far in excess of any readily accessible Zeemann energy..One promising case would be a pair of ions with spins coupled by anisotropic exchange energy (isotropic exchange does not work, since it commutes with the coupling $\vec{s}\cdot\left(\vec{S}_1 + \vec{S}_2\right)$ to the conduction electron spin). For example, an exchange $J(S_{1x}S_{sx} + S_{1y}S_{sy})$ would give an energy gap of order J, commonly of order of several hundred cm^{-1}. If $\mu_B H$ in the foregoing results is replaced by this energy, it would give almost 100% electron spin polarization. (However, there may be a serious problem here: energy conservation obviously requires that the electrons are hot enough to deliver

this kind of energy in the mutual spin flip process. Assuming a mobility of 1000cm/sec/volt/cm, with the translational velocity acquired from the electric field totally randomized, an energy gap of 100 cm^{-1}would require an electric field of about 10^4 volts/cm. Clark and Feher in their experiment noted that a field of only 150 volts/cm already led to breakdown, probably by impact ionization of donor ions.) Finally, implantation may be avoided altogether by building a heterostructure consisting of a thin semiconducting film sandwiched between two antiferromagnetic insulators. If the anisotropy and exchange energies of the latter are J and K respectively, their lowest excitation energy is of order $\sqrt{JK}$,again far above Zeemann energy in a commonly used magnetic field. A full analysis requires allowing for position dependence of $A_\pm$ and $B_\pm$ and excitation of the antiferromagnetic film along the structure. This will be presented in a future calculation.

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